Supplementary Information

Gate tunable spatial accumulation of valley-spin in chemical vapor deposition grown 40°-twisted bilayer WS_2

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Raman spectra, photoluminescence (PL) spectra, and XPS were used to characterize the 40°-twisted bilayer WS_2 samples. As show in Fig. S1, the characteristic vibration peaks E^1_{2g} and A_{1g} of t-WS_2 are obviously visible. Meanwhile, the interlayer exciton peak J_1 (230.1 cm^{-1}), J_2 (296.3 cm^{-1}) and J_3 (322.7 cm^{-1}) is also clearly visible due to the interlayer interaction. However, compared with 0-degree-twisted bilayer WS_2, the characteristic peak of 40°-twisted bilayer WS_2 is notably blue-shifted, which may be due to the difference of interlayer molar exciton interference caused by the change of stacking angle. Moreover, such changes caused by different stacking angles are also reflected in the PL spectra. the characteristic peak of 0°-twisted bilayer WS_2 is significantly redshifted from 1.96 to 1.94 eV compared with that of the 40°-twisted bilayer WS_2, and the peak intensity is significantly weakened. Surprisingly, a new peak appears at 1.98 eV derived from interlayer excitons. At the same time, XPS was used to analyze 40°-twisted bilayer WS_2, and obvious characteristic peaks of S 2p and W 4f were visible, and the stoichiometric ratio of S : W atom is about 2, indicating the crystallinity perfection of the 40°-twisted bilayer WS_2.

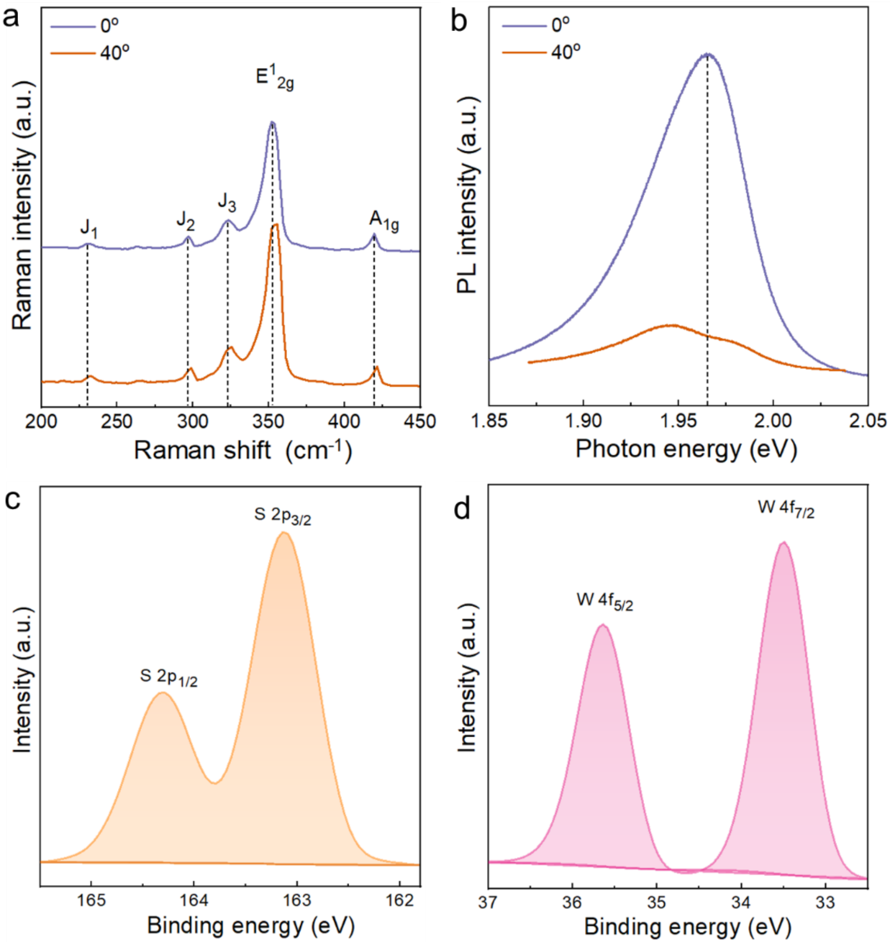

Fig. S1. (a) Raman and (b) PL spectra of 40°-twisted bilayer WS_2 compare to 0°-twisted bilayer WS_2. (c) S 2p and (d) W 4f spectra XPS data of 40°-twisted bilayer WS_2.

The low-magnification STEM diagram of 40° twisted bilayer WS_2 shows in Fig. S2(a). The difference between monolayer and bilayer can be clearly seen. The bright triangle area inside is a bilayer structure, while the outside is a single layer. The atomic structure of monolayer hexagonal crystal system is displayed externally in Fig. S2(b). Meanwhile, STEM image of 0° twisted bilayer WS_2 is also carried out in Fig.

S2(c), and present the AA stacked atomic arrangement structure. The 40°-twisted bilayer WS_2 shows regular Moiré fringe, which proves the existence of Moiré structure in 40° stacked WS_2.

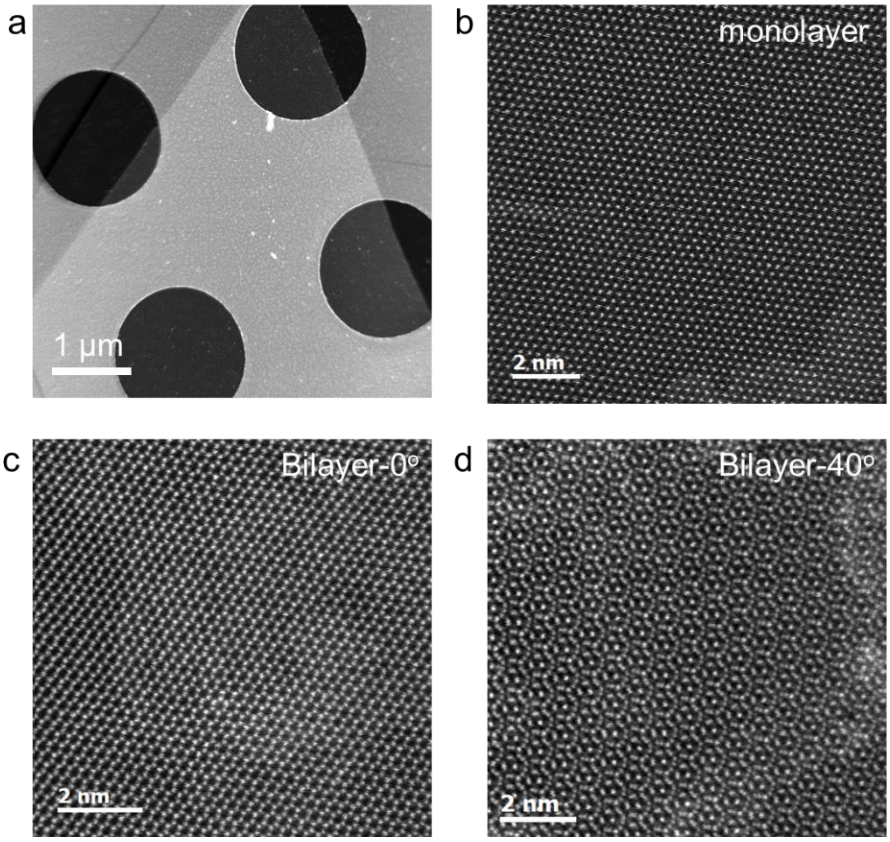

Fig. S2. (a) Low magnification STEM image of 40°-twisted bilayer WS_2. (b) STEM images of monolayer WS_2. STEM images of (c) 0° and (d) 40°-twisted bilayer WS_2.

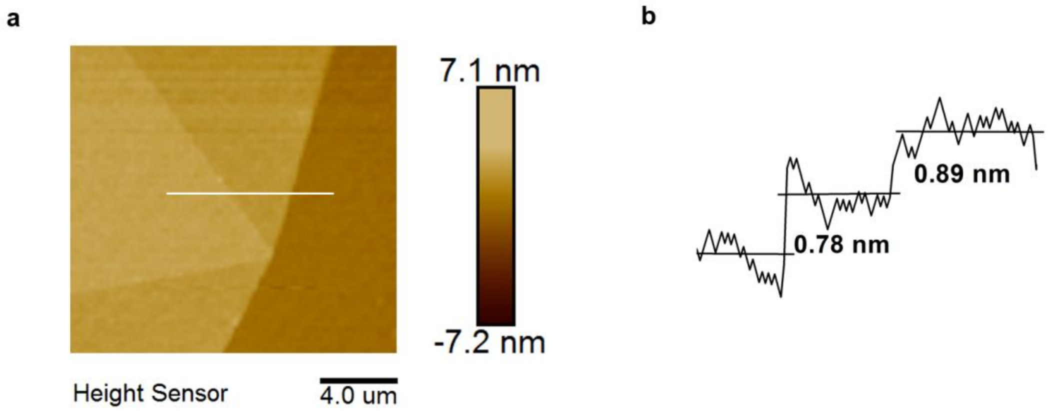

Fig. S3. (a) AFM image of a typical twisted bilayer WS_2. (b) Step height measurement from substrate to the monolayer and bilayer step.